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Date of request ~~[illegible]~~ 2/16/96 — Expected receipt of document 3/~~[illegible]~~16/~~[illegible]~~96

Document number ~~KP-1998~~ K/EM-380 — Date of document 7/13/60

Title and author (if document is unnumbered)

(This section to be completed by Document Center)

Date request received 2/20/96

Date submitted to ADC 2/21/96

Date submitted to HSA Coordinator 2/20/96

(This section to be completed by HSA Coordinator)

Date submitted to CICO 2/21/96 3/13/96 3/19/96

Date received from CICO 2/21/96 3/19/96

Date submitted to ChemRisk/Shonka and DOE 3/25/96

(This section to be completed by ChemRisk/Shonka Research Associates, Inc.)

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2819

K/EM-380

SANITIZED VERSION OF TRACE ELEMENTS EVALUATION (7/13/60)

(SANITIZED VERSION OF CRD DOCUMENT # KP-1998)

Compiled by
S. G. Thornton
Environmental Management Division
OAK RIDGE K-25 SITE
for the Health Studies Agreement

March 13, 1996

Oak Ridge K-25 Site
Oak Ridge, Tennessee 37831-7314
managed by
LOCKHEED MARTIN ENERGY SYSTEMS, INC.
for the U.S. DEPARTMENT OF ENERGY
under Contract DE-AC05-84OR21400

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UNION CARBIDE

INTERNAL CORRESPONDENCE

UNION CARBIDE NUCLEAR COMPANY • ... TENNESSEE

To (Name)	Mr. H. G. P. Snyder	Date	July 13, 1960
Company	Mr. E. C. Bollinger	Originating Dept.	Production Engineering
Location		Answering letter date	
Copy to	Mr. J. Dykstra Mr. K. M. Jones Mr. W. D. McCluen Mr. J. A. Parsons Mr. M. F. Schwenn Mr. S. S. Stief Production and Process Engineering Files	Subject	Trace Elements Evaluation

KP-[redacted]

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The detection of small quantities of impurities within our process equipment and in the process streams has become of increasing importance over the years.

the recovery of some of these materials has also become of interest, and (3) The possibility that these materials may be a health hazard, since most are radioactive, has also been under investigation. Currently, several plant groups are actively investigating these problems with efforts being made to utilize any data obtained for evaluating all problems. The groups investigating the problems are as follows:

Technical Division - Chemical Development

This group has been obtaining samples of barrier at various points throughout the cascade.

A greater part of the data currently available on trace impurities has been obtained from these barrier samples.

Technical Division - Engineering Development

This group has been obtaining samples on cascade streams in an effort to obtain a material balance on trace impurities entering and leaving the cascade. Accountability samples will primarily be used as a source of data. Accountability samples will be sub-sampled in the laboratory and composited for this purpose. In addition, samples of purge gases are being obtained. This group is also working on continuous gas samplers which could be used on the feed stream.

Remarks: CRD
9/27/90

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A second objective of this group is to develop a filter which could be placed in the feed line which would remove the trace impurities before they enter the plant. This work has a dual objective to prevent further contamination of the cascade and to recover the trace material.

Health Physics

As a result of the high alpha radiation associated with neptunium, the determination of accumulations of this element (and any other possible hazardous radiation emitting element) in the cascade and in supporting facilities is being investigated. For this purpose, samples have been obtained throughout the cascade, feed plant, and K-1420. Cascade samples include the barrier samples obtained by Chemical Development, samples of dust residues and surface wipe samples. Feed plant samples were taken at various points in the process and in K-1410 decontamination solutions obtained from feed plant equipment. Samples have also been obtained at various points in the decontamination cycle in K-1420, plus samples of starting material and nickel residue materials returned from INCO.

Process Engineering

Process Engineering has been working with the three above groups in collecting the samples needed to obtain the desired data. Cascade samples, other than the barrier, have been obtained by Process Engineering. Although K-1420 and feed plant samples were taken by operating personnel, arrangements to obtain these samples were made by Process Engineering.

The prime objective of Process Engineering has been to evaluate the plant health hazard problem, if any, created by these trace impurities. Activities have, therefore, been directed toward obtaining samples for this purpose, and to aid in their evaluation.

PROGRESS TO DATE

Analytical data obtained to date are insufficient to obtain any conclusions on any of the objectives in the trace impurity work. However, there are some indications at present which can be used to indicate the course of the investigations in the future. These can best be analyzed by considering each element individually.

Technetium

Data to date indicate a definite gradient of technetium on plant barrier, with the highest concentrations at the reactor tails feed point extending in diminishing quantities upwards through K-33. Very little technetium has been found above K-33. As is shown in table 1, concentrations as great as 31 ppm are present in barrier in K-902-1.

From a health physics standpoint, the possible hazard due to the inhalation of technetium is less severe than uranium, and therefore current limits placed on uranium are also adequate for technetium.

Technetium is present in reactor tails material obtained from Savannah River/ and is present, therefore, throughout all the processing steps in the feed plant. Balances have been run both on our feed plant and on the Paducah feed plant which show that most of the technetium leaves with the uranium hexafluoride produced and is fed to the cascade. The Paducah tests show that approximately 93% of the technetium leaves with the uranium. The Paducah tests also showed that feed cylinder heels become enriched in technetium to some extent, but that this enrichment builds up to a point beyond which all the technetium is fed to the cascade on subsequent feed cycles.

Technetium is also present in nickel residue returned from INCO, indicating the presence of this element in starting material. Analysis of the residue has indicated 14 to 20 ppm in residues from ORGDP and Portsmouth, indicating that this element is not completely removed during decontamination. One batch of residue originating from Paducah and not decontaminated gave a result of 141 ppm. Concentrations in this order would also be possible from residues originating from K-902-1 if the starting material sent to INCO were not decontaminated.

Neptunium

The problem with neptunium is potentially more serious than with technetium, since health physics limitations for neptunium are the same as for plutonium. The current criteria for neptunium, as established in Dr. Henry's letter of May 18, 1960, "Cascade Contaminants," are that if the neptunium content on plant surfaces is such that the neptunium content is greater than 100 ppm based on the uranium present, the plutonium standard would be used. Air counts permissible, therefore, would be reduced from 1 count per minute per cubic foot to 0.1 count per minute per cubic foot. Investigations to date have indicated that neptunium is widespread throughout the cascade (see tables 1, 2, and 3) with no particular gradient, and that the concentrations of neptunium on barrier is well in excess of the 100 ppm. The limited data available to date on other samples taken from the cascade indicate that the neptunium concentration in uranium is below 100 ppm. As a result, it appears that no problem more severe than that caused by uranium is in existence in the cascade field activity since maintenance work does not disturb the barrier. Considerably more data are required to establish this fact more definitely.

The high neptunium to uranium ratio on barrier may present a problem in K-1420, especially in the disassembly of converters and in detubing. Very little data are available to determine whether a problem exists in K-1420, or what happens to the neptunium present on the barrier. Samples have been taken at various points in the decontamination and recovery cycle, but results are not as yet available. In addition, samples of starting material and nickel residue have also been sent to the laboratory for analysis.

One sample of nickel residue, originating from Paducah starting material, has been analyzed. The neptunium concentration was 0.52 ppm.

No neptunium problem seems to exist in the feed plant at present. The neptunium concentration in Savannah River oxides is in the order of a few parts per billion. This indicates that the neptunium currently in the cascade results from Savannah River material which was fed several years ago when the concentration was appreciable, or comes from Paducah in Paducah product feed.

Strontium

Very small amounts of strontium have been found at two points in the cascade (see table 1). No problem from this element appears to exist in the cascade; however, the source of the strontium is not evident since the points at which strontium was found are remote from the feed point. More data are required to evaluate the problem. The laboratory is currently working on a procedure to lower the limit of detectability of this material.

Somewhat larger accumulations of strontium have been found in filters at the feed plant. Here again, very little data are available (1 or two samples) and, as a result, no evaluation can be made. Samples from the feed plant are currently being analyzed to obtain more information.

Plutonium

The plutonium analyses obtained to date are all <1 ppb. No problem appears to exist from plutonium.

Thorium, Ruthenium, and Others

Several small concentrations of ruthenium have been found on barrier removed from K-33 (see table 1). Small concentrations of thorium have been found on almost all barrier analyzed. Normal precautions necessary to provide protection against uranium will also be adequate for these beta-gamma emitters. No problem appears to exist from this source in the cascade. Very little data are available for these elements at the feed plant; however, thorium, ruthenium, and some others have been detected at this point.

FUTURE PLANS

More emphasis will be placed on investigating potential health physics problems in the K-1420 building and in the feed plant, since there is a greater likelihood of a problem at these points than in the cascade. A problem requiring immediate attention in K-1420 is the concentrations of trace impurities in the starting material and their possible impact on the INCO contract. Samples will also be taken in the cascade whenever maintenance activity permits the obtaining of samples from points not previously covered.

Engineering Development has accumulated 180 samples covering cascade streams for the period April 10 to May 10 (the tenth of the month was chosen to prevent conflict with the laboratory sample load originating from the monthly cascade inventory). These samples will be composited and analyzed to attempt a material balance for the trace impurities. Test adsorbers will also be installed in the K-33 feed line to test materials which would remove trace impurities and possibly permit recovery. Sodium fluoride will probably first be tested since it is known the molybdenum, vanadium, and technetium will adsorb on this material to some extent.

Engineering Development will also continue work on automatic gas sampling systems for the K-33 feed line to facilitate the obtaining of more representative samples.

Works Laboratory Participation

At the present time, the limiting factor to the trace impurity investigation is the capacity of the Works Laboratory. Currently, six people are spending full time in the radiochemical analysis of the samples with a maximum capacity estimated at 30 samples per week. To date, 224 samples have been obtained from the cascade in addition to barrier samples. Some compositing has been necessary to reduce the number of analyses. The program envisioned by Engineering Development entails approximately 30 samples in the laboratory from K-1420 and K-1131. Obviously, samples can be obtained much faster than they can be analyzed. As a result, it appears that the program becomes fairly long-term in nature in order to collect sufficient data to obtain the desired results. Efforts will be made to reduce the sample load whenever possible.

Currently, radiochemical analyses are being charged to account No. 1340-45. The estimated cost is in the order of $60,000 per year (including plant expense).

S. S. Ste
for J. A. Parsons

SSS/eb

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TABLE 1

CONCENTRATION OF TRACE IMPURITIES ON BARRIER SAMPLES

Location	Thorium $\mu g./g. \times 10^{-8}$	Neptunium $\mu g./g.$	Technetium $\mu g./g.$	Strontium $\mu g./g. \times 10^{-6}$	Ruthenium $\mu g./g. \times 10^{-8}$	Plutonium ppb.
K-502-3.6.2-3-4	1.1	0.510	<0.02	<1	<1	<1
-7	0.8	0.348	<0.02	<1	<1	<1
K-602-2.6.3-2-1	4.5	0.209	<0.02	<1	<1	<1
-4	4.1	0.313	<0.02	<1	<1	<1
.6.6-2-1	18.2	<0.010	<0.02	<1	<1	<1
-7	15.9	0.109	0.08	<1	<1	<1
.6.8-1-4	4.5	0.168	<0.02	<1	<1	<1
-7	4.7	0.204	0.04	<1	<1	<1
.6.9-1-1	3.4	<0.010	0.48	<1	<1	<1
-7	14.1	0.309	0.04	<1	<1	<1
K-602-3.4.1-1-1	1.4	0.332	<0.02	<1	<1	<1
-4	2.3	0.242	<0.02	<1	<1	<1
.6.6-3-4	0.8	0.162	<0.02	<1	<1	<1
-7	1.5	0.324	<0.02	<1	<1	<1
.8.7-1-1	5.2	0.197	<0.02	<1	<1	<1
-7	26.9	0.067	<0.02	<1	<1	<1
K-602-4.4.5-1-1	0.5	0.357	<0.02	<1	<1	<1
-4	1.0	0.244	<0.02	<1	<1	<1
.9.1-1-4	4.4	0.130	<0.02	<1	<1	<1
-7	7.3	0.170	<0.02	1.6	<1	<1
K-902-1.10.7-B-1	2.8	0.531	26.51	<1		<1
-B-7	17.7	0.277	21.13	<1		<1
K-902-2.4.2-2-1	2.8	0.309	7.83	<1		<1
-4	3.7	0.535	6.58	<1		<1
K-902-5.2.3-2-4	4.2	0.248	4.3	<1	24.5	<1
-7	3.5	0.140	7.1	<1	13.0	<1
K-902-6.7.3-3-1	1.7	0.155	1.3	2.3	20.7	<1
-7	1.7	0.399	1.1	<1	13.0	<1
K-902-8.9.3-3-1	5.2	0.467	4.7	<1		<1
-4	1.7	0.120	5.5	<1		<1

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TABLE 2

CONCENTRATION OF TRACE IMPURITIES IN CASCADE SAMPLES

		g. U per gram	Thorium Beta Dis. per g.	Neptunium µg./g.	Technetium µg./g.	Strontium µg./g. x 10^{-6}	Plutonium µg./g. x 10^{-3}	Total Alpha Counts/g.	Total Beta Dis./g.	µg. Np per g.U
K-902-8.9.1	TNR-1	0.598	1,074,134	0.522	1.31	<1	<2	282,090	888,858	0.87
	TNR-2	0.581	1,004,290	0.313	1.08	<1	<2	306,662	873,932	0.54
	TNR-3	0.678	1,072,860	1.13	1.12	<1	<2	351,842	1,135,428	1.7
K-602-1.1.8	TNR-1	0.633	610,216	0.954	0.26	<1	<2	246,900	585,800	1.5
K-902-2.3	TNR-1	0.676	880,666	0.858	<0.02	<1	<2	304,442	1,123,203	1.3
K-602-2.7.10	TNR-3	0.537	775,760	0.328	5.03	<1	<2	246,148	1,212,927	0.61
K-902-2.3	TNR-7	0.606	851,443	<0.010	0.94	<1	<2	217,463	873,010	<0.02
K-502-1.9.9	TNR-29	0.378	1,254, 59	<0.010	20.18	<1	<2	856,995	1,329,176	<0.03
K-902-8.9	3 CS	0.0511	175,969	1.63	101.9	<1	<2	28,790	2,267,617	32.
K-402-7.9	TNR-24	0.269	1,575,850	<0.010	<0.02	<1	<2	1,462,120	1,092,960	<0.04
K-502-1.10	SCV	0.424	817,658	<0.010	<0.02	<1	<2	352,540	1,175,476	<0.02
K-303-5.1	TNR-6	0.353	3,291,545	<0.010	<0.02	<1	<2	5,199,500	3,749,110	<0.03

		g. U per Sample	Thorium Total Beta Dis.	Neptunium µg./Sample	Technetium µg./Sample	Strontium µg. x 10^{-6} per Sample	Plutonium µg. x 10^{-3} per Sample	Total Alpha Counts	Total Beta Dis.	µg. Np per g. U
K-902-8.9.1	B	0.4193	120,480	0.886	2.48	<1	<2	43,000	1,019,500	2.1
K-502-1.9	ID	0.0471	1,956,680	<0.010	<0.02	<1	<2	13,000	4,513,000	<0.2
K-402-7.10	CV F.P.Wipes	0.0072	2,238,760	<0.010	<0.02	<1	<2	45,000	2,397,500	<1.4
PPU-FP		0.0178	611,868	<0.010	<0.02	<1	<2	719,500	981,500	<0.6
K-303-5.1	I TNW Inlet	0.0044	340,208	<0.010	<0.02	<1	<2	88,000	472,850	<2.
K-402-7.9	CP 8 F.P.Wipes	0.0012	63,080	<0.010	<0.02	<1	<2	750	485,825	<10.
K-602-1.1.8	CF	*	301,900	<0.010	<0.02	<1	<2	52,000	433,675	-
PPU-PR	TNR-13	0.0014	37,750	0.190	<0.02	<1	<2	27,800	54,300	140.
K-902-2.3	C	0.0111	11,896	0.709	0.21	<1	<2	5,454	32,340	64.
K-602-2.7.10	C	0.0114	374,912	0.705	6.82	<1	<2	5,360	719,600	62.
PPU-F		0.1390	3,810,640	<0.010	<0.02	<1	<2	7,199,000	7,617,000	<0.1

* Operational error.

TABLE 3

CONCENTRATION OF TRACE IMPURITIES IN FEED PLANT SAMPLES

Sample Number	g.U/g.	Thorium μg. x 10^{-6}/g.	Neptunium μg./g.	Technetium μg./g.	Strontium μg. x 10^{-6}/g.	Plutonium ppb/U	Total Beta Dis./g.	Total Alpha Cts./g.	Gamma Activity, % (As to Natural Uranium)	μg. Np per G.U
HPX-601 (K-1302 Stock Material)	0.607	700	<0.010	<0.02	<0.5	<2	975,980	578,045	179	< 0.02
HPX-101 (Enriched UO_3)	0.804	928	0.018	0.99	<0.5	3	1,025,425	250,940	168	< 0.02
HPX-101A (Y-12 UO_3)	0.818	1,148	-	<0.02	<0.5	<2	1,018,954	238,552	100	-
HPX-102 (UO_2)	0.875	1,224	0.024	1.28	<0.5	<2	1,122,148	272,800	100	0.03
HPX-103 (UF_4)	0.765	1,038	<0.010	0.97	<0.5	<2	972,906	211,030	100	< 0.02
HPX-104 (No. 3 Tower Ash)	0.733	2,210	0.041	0.10	35.1*	3	1,973,677	231,832	232	0.06
HPX-105 (No. 3 Cyclone Ash)	0.033	107,000	0.037	64.96	494.7*	2,199	105,876,260	206,778	437,600	1.0
HPX-106 (No. 3 Barrier Ash)	0.209	121,132	0.030	10.49	538.4	1,452	132,446,610	247,376	83,000	0.14
HPX-107 (No. 5 Tower Ash Cleanup Reactor)	0.746	1,415	0.039	0.49	13.1	4	1,912,329	241,096	304	0.05
HPX-113 (UO_3)	0.827	787	0.041	10.48	<0.5	<2	807,846	146,462	194	0.05
HPX-114 (UO_2)	0.863	619	0.024	10.10	<0.5	<2	996,930	197,895	159	0.03
HPX-115 (UF_4)	0.763	863	<0.010	0.34	<0.5	<2	902,500	180,740	100	< 0.02
HPX-116 (No. 3 Tower Ash)	0.726	836	<0.010	0.34	<0.5	<2	751,077	214,769	100	< 0.02
HPX-117 (No. 4 Cyclone Ash)	0.113	127,668	0.138	45.3	640	1,015	128,015,000	441,203	17,700	1.2
HPX-118 (No. 4 Barrier Ash)	0.200	287,162	0.024	133.89	473	861	235,142,676	259,634	426,800	0.12
HPX-119 (Cleanup Reactor Ash)	0.719	3,059	<0.010	4.27	-	13	1,008,400	217,174	700	< 0.02
HPX-121 (Filter Paper)	-	5	0.139	<0.02	<0.5	-	3,600	1,400	N. D.**	-
HPX-123 (Filter Paper)	-	5	<0.010	<0.02	<0.5	-	25,800	1,700	N. D.**	-
N-17 (Nickel Residue)	-	215	<0.010	17.08	-	-	-	-	-	-

* Corrected results.

** None detected.

TABLE 4

CONCENTRATION OF TRACE IMPURITIES IN STARTING MATERIAL
AND INCO RESIDUE

	Sample Number	gu/g.	Per Cent U-235	Neptunium µg./g.	Technetium µg./g.	Plutonium ppb	Ruthenium µg./g.	Strontium µg./g.
INCO Residue	C-201	0.010	0.560	<0.01	-	-	-	-
	K-206	0.0048	0.856	<0.01	12.88	-	-	-
	Y-209	0.0034	0.784	<0.01	12.06	-	-	-
	C-207	0.0178	0.406	0.52	141	<1	2.2×10^{-7}	1.1×10^{-6}
	C-205	-	-	0.336	23.26	-	-	-
	C-210	-	-	0.256	71.13	-	-	-
Starting Material	224A	0.000063	-	<0.01	<0.02	-	-	-
	224B	0.000049	-	<0.01	<0.02	-	-	-

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